PRODUCTION OF SAMPLES OF INDIVIDUAL RADIOXENON ISOTOPES THROUGH NEUTRON IRRADIATION OF STABLE XENON GAS

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Sponsored by Army Space and Missile Defense Command

Contract No. W9113M-05-1-0017
Proposal No. BAA05-49

ABSTRACT

The Spectral Deconvolution Analysis Tool (SDAT) software was developed to improve counting statistics and detection limits for nuclear explosion radionuclide measurements. SDAT utilizes spectral deconvolution spectroscopy techniques and can analyze both β-γ coincidence spectra for radioxenon isotopes and high-resolution HPGe spectra from aerosol monitors.

The deconvolution algorithm of the SDAT requires a library of β-γ coincidence spectra of individual radioxenon isotopes to determine isotopic ratios in a sample. In order to get experimentally produced spectra of the individual isotopes, we have irradiated enriched samples of ^{130}Xe, ^{132}Xe, and ^{134}Xe gas with a neutron beam from the TRIGA reactor at The University of Texas. The samples produced were counted in an Automated Radioxenon Sampler/Analyzer (ARSA) style β-γ coincidence detector. The spectra produced show that this method of radioxenon production yields samples with very high purity of the individual isotopes for ^{131m}Xe and ^{135}Xe and a sample with a substantial ^{133m}Xe to ^{133}Xe ratio.

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE **SEP 2008**	2. REPORT TYPE	3. DATES COVERED **00-00-2008 to 00-00-2008**

4. TITLE AND SUBTITLE **Production of Samples of Individual Radioxenon Isotopes Through Neutron Irradiation of Stable Xenon Gas**	5a. CONTRACT NUMBER
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Pacific Northwest National Laboratory,PO Box 999,Richland,WA,99352**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES
Proceedings of the 30th Monitoring Research Review: Ground-Based Nuclear Explosion Monitoring Technologies, 23-25 Sep 2008, Portsmouth, VA sponsored by the National Nuclear Security Administration (NNSA) and the Air Force Research Laboratory (AFRL)

14. ABSTRACT
see report

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**	**Same as Report (SAR)**	**9**	

OBJECTIVES

The goal of the experiment is to produce β-γ coincidence spectra of gaseous samples of each radioxenon isotope of interest in nuclear explosion monitoring. This experiment will take a new approach to creating radioxenon through neutron irradiation of stable xenon gas. The data is needed as a required library data set for the operation of the SDAT (Foltz Biegalski 2003, Biegalski 2005).

In order to produce the radioxenon isotopes of interest ^{130}Xe must be activated to ^{131m}Xe, ^{132}Xe must be activated to both ^{133}Xe and ^{133m}Xe, and ^{134}Xe must be activated to ^{135}Xe. The irradiation is done in Beam Port 2 of The University of Texas at Austin 1.1 MW TRIGA reactor. The beam port is tangential to the core and has a neutron collimator. The thermal neutron equivalent flux emerging from the beam port is 1.24 x10^8 n cm^{-2} s^{-1} when the reactor is operating at 950 kW (Whitney 2006).

The gas is then transferred to a detector system very similar to that in the ARSA for counting (Bowyer 1999). The β-γ coincidence spectra that are produced are analyzed for purity and the effectiveness of the procedure is determined. For a more detailed explanation of the work, see the dissertation by Haas referenced here (Haas 2008).

RESEARCH ACCOMPLISHED

Results of ^{130}Xe Irradiation

The production of ^{131m}Xe through irradiation of enriched ^{130}Xe is shown in Figure 1. The spectrum initially contained noticeable amounts of ^{133}Xe and ^{135}Xe. After three days of decay, the activity of ^{131m}Xe has been reduced by less than half, but this constitutes more than seven half lives of ^{135}Xe, effectively reducing it to background levels.

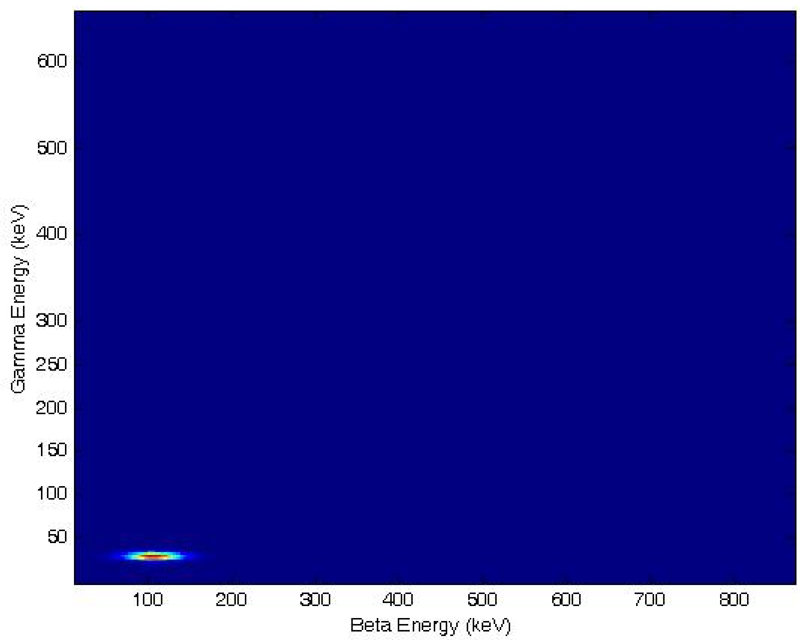

Figure 1. Spectrum of a sample containing only ^{131m}Xe—6.9-hour irradiation, 73.4-hour decay, 87.6-hour count

The data in Figure 1 are analyzed and the initial ratio of ^{131m}Xe/^{133}Xe by activity at the time the sample is placed in the detector is found to be 316, or a sample that is 99.7% ^{131m}Xe by activity. If greater purity is needed, the sample can be allowed to decay further to increase the ratio, but this will also increase the effect of background counts on the sample.

Results of ^{132}Xe Irradiation

The irradiation of ^{132}Xe results in both ^{133}Xe and ^{133m}Xe. Because ^{133m}Xe has a shorter half life than ^{133}Xe the optimum ratio of ^{133m}Xe to ^{133}Xe occurs immediately after irradiation. Figure 2 shows the spectrum produced when both ^{133m}Xe and ^{133}Xe are present.

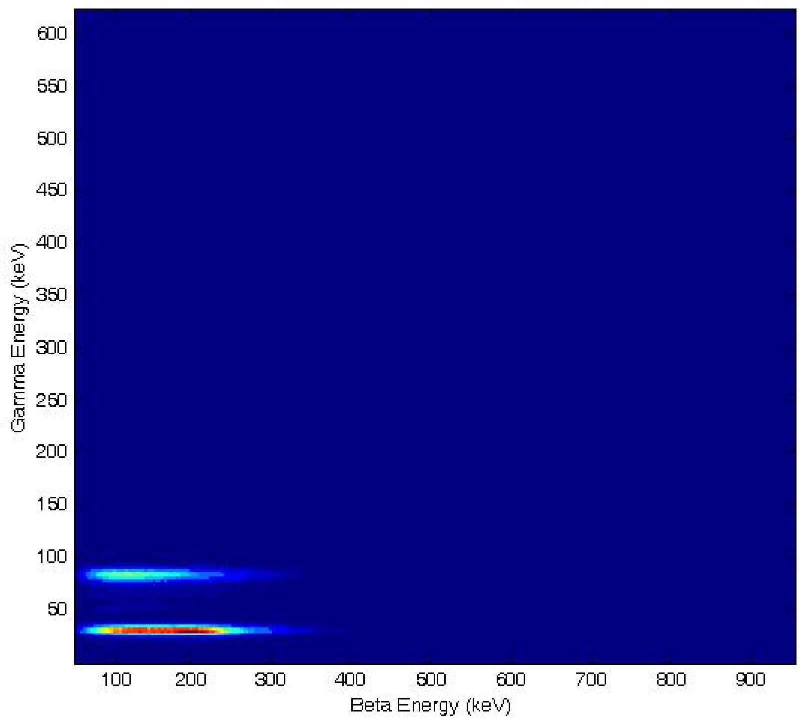

Figure 2. Spectrum of a sample containing both ^{133}Xe and ^{133m}Xe—7.3-hour irradiation, 1.5-hour decay, 209.6-hour count.

Figure 3 is a surface plot of the same data as Figure 2 and more obviously depicts the ^{133m}Xe peak. Due to the greater probability that a ^{132}Xe nucleus will be activated to the ground state than the metastable state of ^{133}Xe, ^{133m}Xe cannot be produced without simultaneously producing a greater activity of ^{133}Xe using this method of radioxenon production.

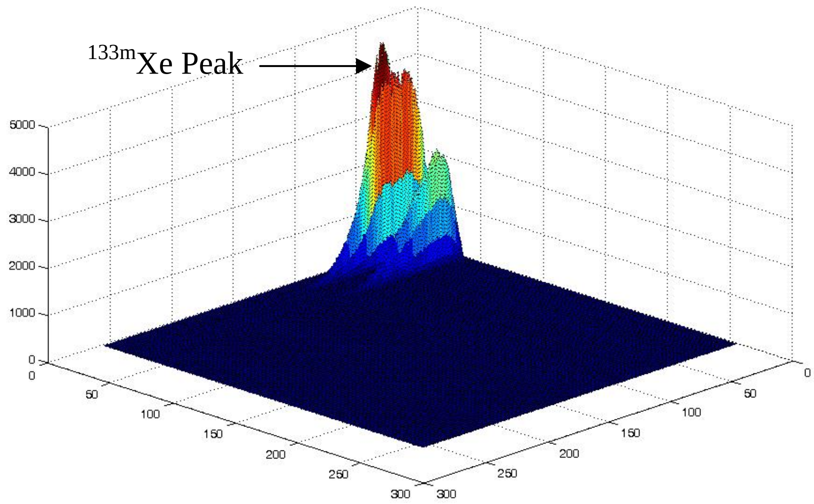

Figure 3. Alternate view of mixed-sample spectrum—7.3-hour irradiation, 1.5-hour decay, 209.6-hour count.

The procedure for this sample is to collect one mixed spectrum containing ^{133m}Xe and ^{133}Xe and one spectrum of the remaining ^{133}Xe after the ^{133m}Xe has decayed to background levels. A spectrum of ^{133}Xe normalized to match the counts of ^{133}Xe in the mixed spectrum can be subtracted from the mixed spectrum to yield a spectrum containing only ^{133m}Xe.

Figure 4 shows the spectrum of ^{133}Xe after the ^{133m}Xe has decayed away. The count that produced this spectrum was started 9 days after the sample was produced. Since the expected initial activity of ^{133m}Xe is around 25% of the expected initial activity of ^{133}Xe, the activity of ^{133m}Xe should be only a few percent that of ^{133}Xe after four half lives. Also, the percentage of ^{133m}Xe in the sample will decrease as the count is taken. Figure 5 shows a surface plot of the spectrum of ^{133}Xe where the ^{133m}Xe peak is visibly absent.

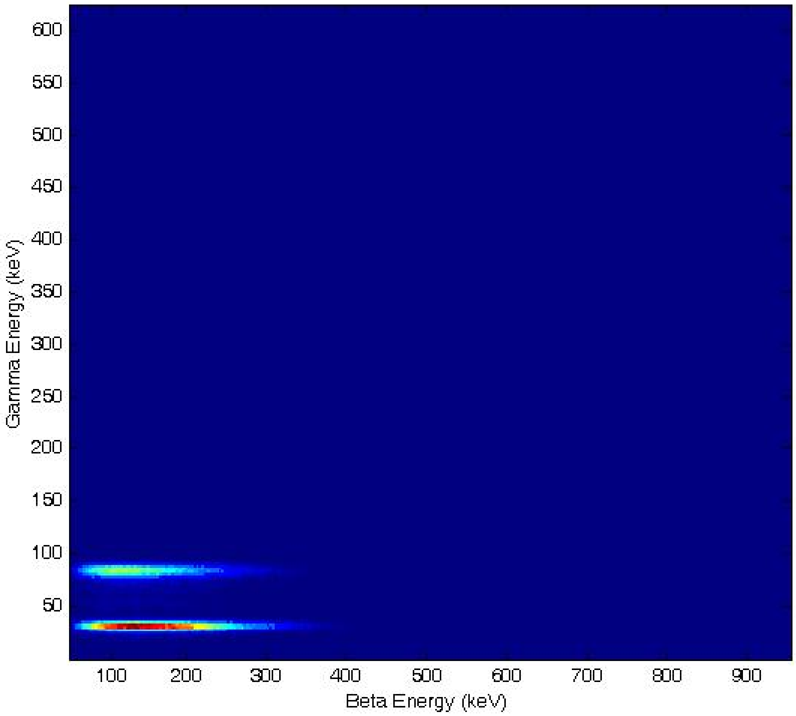

Figure 4. Spectrum of a sample containing high purity ^{133}Xe—7.3-hour irradiation, 211.1-hour decay, 330.2-hour count.

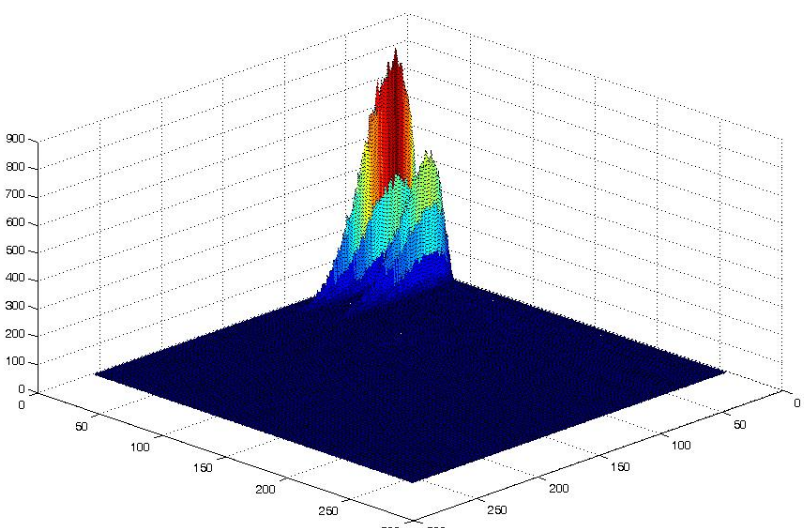

Figure 5. Alternate view of ^{133}Xe spectrum—7.3-hour irradiation, 211.1 hour decay, 330.2-hour count.

The spectrum of ^{133}Xe is normalized so that the 81 keV γ-peak has the same magnitude as the same peak in the mixed spectrum. Then the ^{133}Xe spectrum is subtracted from the mixed spectrum, resulting in a sample consisting solely of ^{133m}Xe. The result is shown in Figure 6.

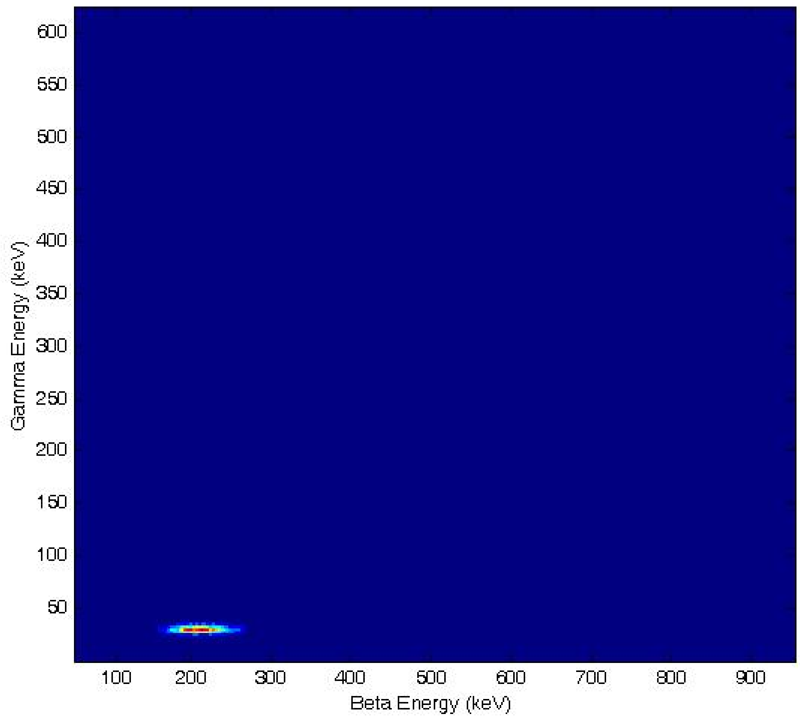

Figure 6. Sample of ^{133m}Xe resulting from subtraction of ^{133}Xe from the mixed sample—7.3-hour irradiation, 1.5-hour decay, 209.6-hour count.

The data in Figures 2 and 6 are analyzed to get the initial activity of each isotope. The 81 keV peak data is used to determine the activity of ^{133}Xe. Of particular interest in evaluating the method of neutron irradiation of stable xenon gas is the ratio of ^{133m}Xe to ^{133}Xe that can be produced. The ratio found using an eight-hour irradiation is 0.31 by activity. This corresponds to a sample consisting of 23.6% ^{133m}Xe and 76.4% ^{133}Xe by activity.

Results of ^{134}Xe Irradiation

The most significant results of the enriched xenon irradiation are the spectra of isolated ^{135}Xe. Because ^{135}Xe has the shortest half life of the four radioxenon isotopes of interest, the spectra of ^{135}Xe produced through the fission of uranium have an impurity of the other radioxenon isotopes. The spectrum produced through the irradiation of enriched ^{134}Xe is shown in Figure 7.

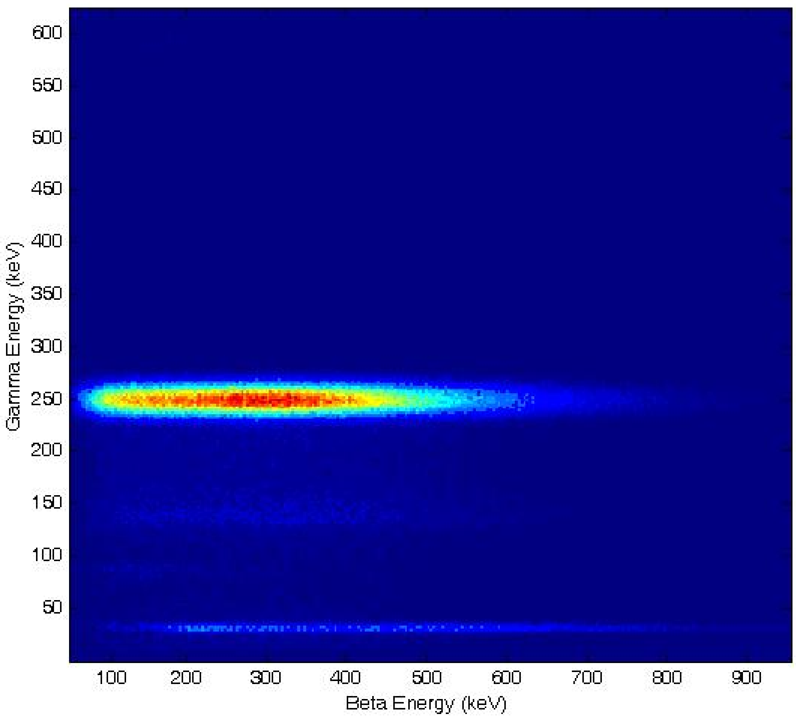

Figure 7. Experimental ^{135}Xe spectrum with trace amounts of ^{133m}Xe and 133Xe—6.9-hour irradiation, 0.6-hour decay, 18.6-hour count

The data from Figure 7 are analyzed with a longer count taken to provide more ^{133}Xe counts. The 250 keV peak and 81 keV peak data were used to determine the activity of ^{135}Xe and ^{133}Xe, respectively. The same calculation can be done for the ratio of ^{135}Xe to (^{133}Xe+^{133m}Xe) as was done for the ratio of ^{133m}Xe to ^{133}Xe. The sample of irradiated enriched ^{134}Xe is found to be 99.1% ^{135}Xe by activity when the sample is placed in the detector.

Analysis of the Experimental Method in Context

The purpose for conducting these experiments was to produce spectra of the radioxenon isotopes that are of greater isotopic purity than those that have been produced in the past. In order to compare this method to others, we must find the activity of each isotope in each sample. Using the efficiency calculations from modeling and the counts-per-second data from the experiment section, we can determine the activity of the samples produced.

The sample of isolated ^{131m}Xe is not as significant as the samples of the other isotopes due to the ability to wait for any sample of mixed radioxenon gas to decay until only ^{131m}Xe remains. These data do prove, however, that the neutron irradiation of stable ^{130}Xe can be used to produce samples of ^{131m}Xe.

In order to compare this method to radioxenon production through the fission of ^{235}U, we can derive the ratios of samples that can be extracted from the fission gases. The direct fission yields of ^{235}U show that isotopic ratios greater than those achieved through stable gas irradiation are possible with this method, but the build-in of ^{133}Xe occurs quickly through the decay of ^{133}I.

The production of radioxenon through the fission of ^{235}U is modeled in ORIGEN-ARP2 to determine isotopic ratios from various reactor operation times (ORNL 2004). The results for the ratio of ^{133m}Xe to ^{133}Xe produced through fast separation of fission products are shown in Table 1 with the results from the stable gas irradiation experiment. Because separation is not a factor in the irradiation of stable gas, the ratio as a function of separation time is constant.

Table 1. Initial Ratio by Activity of ^{133m}Xe to ^{133}Xe using Various Methods of Production

^{133m}Xe/^{133}Xe Ratio	Irradiation time (minutes)				Ratio given by
Separation Time (hours)	60	15	5	1	Xe Irradiation Method
0.5	0.115	0.164	0.186	0.196	
1	0.096	0.108	0.113	0.114	
3	0.078	0.080	0.080	0.080	0.31
5	0.074	0.075	0.075	0.075	
10	0.071	0.071	0.071	0.071	

The table above shows that even with a very short irradiation time, the ratio is not as high for the ^{235}U method as for the xenon irradiation method. This calculation is also done to determine the ratio of ^{135}Xe to the other radioxenon isotopes. This is shown in Table 2.

Table 2. Initial ratio by activity of ^{135}Xe to other radioxenon isotopes using various methods of production

^{135}Xe/(^{133}Xe+^{133m}Xe+^{131m}Xe) Ratio	Irradiation time (minutes)				Ratio given by
Separation Time (hours)	60	15	5	1	Xe Irradiation Method
0.5	85	127	140	143	
1	67	84	88	89	
3	42	46	46	46	106
5	33	34	35	35	
10	21	22	22	22	

The two tables above show that the fast separation of the radioxenon isotopes may provide a method for achieving a higher initial ratio of the activity of ^{135}Xe to the activities of the other radioxenon isotopes, but its application to the production of pure ^{133m}Xe may be limited. The tables also show that the isotopic ratios produced using the irradiation of enriched stable xenon gas are significantly higher than those produced by systems that take hours to separate the radioxenon from other fission products.

CONCLUSIONS AND RECOMMENDATIONS

The experiment to produce β-γ coincidence spectra of individual radioxenon isotopes through the neutron irradiation of enriched stable xenon gas has succeeded. Two of the four isotopes (^{131m}Xe and ^{135}Xe) have been detected with greater than 99% purity. The production of an isolated sample of ^{133m}Xe is not possible using these techniques, but a spectrum of the isotope has been produced through the subtraction of ^{133}Xe. And a spectrum of ^{133}Xe has been collected with trace amounts of ^{133m}Xe and ^{131m}Xe.

The data produced in this work will be used to further the field of underground nuclear explosion monitoring through the SDAT program. The application of this technique will allow the benchmarking of future detectors with samples of individual radioxenon isotopes as opposed to a mixture of all four. These benchmarks, along with improved data-analysis programs such as the SDAT will improve the sensitivity of global monitoring systems.

ACKNOWLEDGEMENTS

Thanks to Ted Bowyer, Justin McIntyre, and Matt Cooper of Pacific Northwest National Laboratory for their instruction on the setup and use of an ARSA style detector.

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